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PHYSICAL REVIEW B VOLUME 44, NUMBER 9 1 SEPTEMBER 1991-I

Observation of an extended region of magnetic reversibility in Nb and $NbSe_2$

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(Received 14 May 1991)

Vibrating-reed measurements of the flexural resonances of thin cantilevered samples of polycrystalline Nb foils and quasi-two-dimensional $NbSe_2$ single crystals reveal extended regions of temperature T and magnetic field H for reversible flux-line (FL) motion when the long reed dimension is perpendicular to the applied field. An "irreversibility line," defined by the peak temperature of the reciprocal quality factor, is found to be frequency (mode) -independent for resonant frequencies $5\times10^2 < f < 5\times10^4$ Hz, implying that the time scale of FL pinning is extremely T and H dependent. We conclude that observation of an extended region of magnetic reversibility is not restricted to high-T_c or extremely anisotropic materials, and depends upon the geometric aspect of samples with respect to the applied field direction.

The discovery of high-superconducting-transition temperatures in the copper oxides (HTO) has renewed interest in the problems of motion and pinning of flux lines (FL) in type-II superconductors. A variety of phenomena occur at temperatures significantly below the equilibrium transition temperature T_c in the HTO, such as extended regimes of reversible magnetization,[1] low critical currents,[2] and damping peaks and frequency shifts of mechanical resonances.[3,4] These results have been variously interpreted in terms of FL melting,[3] FL depinning,[4] and FL glass formation.[5] Different "irreversibility" temperatures observed in different measurements have been associated with different measurement frequencies.[6] These effects, not previously observed in low-temperature superconductors, were thought to arise in HTO as a combined result of their quasi-two-dimensionality (i.e., extremely short coherence lengths ξ_0 perpendicular to the copper planes) and thermally activated FL motion near their very high transition temperatures.[6,7]

Here, we report on elastic ("vibrating reed") and electrical resistivity measurements on polycrystalline Nb foils ($T_c[H=0]=9.2$ K) and quasi-two-dimensional $NbSe_2$ single crystals ($T_c[H=0]=7.0$ K). In spite of the low T_c's and long coherence lengths (430 Å and 26–76 Å, respectively[8,9]) of these materials compared to the HTO, we find an extended region of temperature and field over which reversible FL motion occurs for sample reeds of both Nb and $NbSe_2$. Specifically, in the case of a Nb reed oriented with long dimension perpendicular to the applied field, we observe a strong, frequency (mode) -independent depression of the "irreversibility temperature" $T_Q(H)$ (defined as the temperature of the peak in damping) below the resistively determined critical temperature T_R. We do not find such a depression for the parallel reed orientation in Nb; and we observe a definite, but smaller depression of T_Q for $NbSe_2$ in the perpendicular orientation.

These results are comparable to the recent observations[10] of reversible magnetization over a finite temperature interval below $T_c(H)$ in filamentary Nb-Ti and Nb_3Sn wires by Suenaga, Ghosh, Xu, and Welsh (SGXW). Our results on Nb and $NbSe_2$, and the fact that the superconducting filaments of SGXW were oriented perpendicular to the applied field, strongly suggest that the observation of magnetic reversibility depends on the geometric aspect of the sample with respect to the applied field direction. Further, we conclude that a variety of superconductors, including isotropic, low-T_c materials, can, under suitable conditions, display extended regions of magnetic reversibility with strong mobile vortex effects.

Vibrating reed experiments measure the resonant frequencies and damping of flexural resonances excited in thin, cantilevered samples, and are very sensitive probes of vortex dynamics in superconductors. Brandt and coworkers[11–14] have demonstrated that strong resonant frequency shifts and damping peaks can be observed at a temperature at which the FL become pinned to the crystal lattice. In the present work, we have used the accessibility of higher order overtones (extending from 5×10^2 to 5×10^4 Hz) to probe frequency-dependent behavior of vortex dynamics as a further check on contemporary theories of irreversibility anomalies.

$NbSe_2$ single-crystals were grown by standard vapor transport techniques.[15] The crystals have micaceous morphologies, with quasi-two-dimensional hexagonal *ab* planes weakly bonded via van der Waals interactions, resulting in electronic effective-mass anisotropy ratios $m_c/m_{ab}\approx9$ and coherence length anisotropy $\xi_0(ab)/\xi_0(c)\approx3$ (Refs. 9 and 16). The Nb foil sample was prepared by cold-rolling 99.8% pure ingot. The residual resistivity ratios of the $NbSe_2$ crystals were typically $RRR\approx25$, and the zero-field T_c and transition width ΔT_c had values of 7.0 K and <0.1 K, respectively. The Nb foils exhibited $RRR\approx23$, $T_c\approx9.2$ K, and $\Delta T_c\approx0.07$ K.

Sample dimensions were typically 1 cm×1 mm×50 μm, and one end was epoxied to a piezoelectric transducer used to excite the flexural resonances. The resonant frequency was measured using phase sensitive demodulation[17] of the output of an rf helical resonator detector;[18] typically several resonant modes with frequencies ranging from 500 Hz to 50 kHz could be observed and measured. Mechanical damping was characterized in terms of the reciprocal quality factor, $1/Q$, of the resonance, usually found by monitoring the amplitude of the resonance and assuming that, for a given driving force, the amplitude was proportional to the Q.[17] As emphasized in Refs. 4 and 11, this can be a poor choice for quantitative measurements of the damping because for $T < T_Q$, the damping is itself amplitude dependent. However, in this work, we concentrate on the field dependence of T_Q, which does not vary significantly with amplitude (i.e., $\Delta T_Q < 0.01$ K if self-heating is eliminated). Typical vibrational amplitudes were ≈ 10 Å at the damping peaks. Samples were visually aligned (to approximately a 5° accuracy) parallel or perpendicular to the applied field direction.

Electrical leads were attached using silver epoxy, and four-lead resistivity measurements were carried out using a lock-in amplifier with phase sensitive detector, operating at 14 Hz with 10 μA to 1 mA rms measuring currents.

The electrical resistance R, resonant frequency shift $\Delta f/f$, and internal friction $(1/Q)$ of the fundamental resonance (f = 680 Hz) of a $NbSe_2$ reed are shown as functions of temperature in Fig. 1. The crystal was oriented with the *ab* plane perpendicular (reed $\perp$) to the applied field H = 11 kOe. The resonant frequency increases rapidly at T_Q, where the damping has a sharp maximum. Strong hysteresis is seen in $\Delta f/f$ only below T_Q, justifying its identification with the irreversibility temperature. There are nonhysteretic high-temperature tails in the damping and resonant frequency shifts extending up to the center of the resistive transition. In turn, there is a finite low-temperature tail in the resistance, which only reaches zero at $T_R \approx T_Q$. If one interprets T_Q as the onset of FL pinning,[4,14] the low-temperature, field-sensitive resistive tail is indicative of a region of reversible FL motion. For the parallel orientation, the resistive transition is much sharper and a clear reversible region cannot be resolved in our data. This picture does not preclude a lower temperature region of activated flux creep that should be observed for sample resistance near zero.[7]

The slope of the "irreversibility line" (perpendicular orientation), $dH/dT_Q = -7.6 \pm 0.5$ kOe/K, where the uncertainty reflects sample dependence. This value is consistent with ac susceptibility measurements performed at 1 kHz (-7.3 kOe/K) (Ref. 16) and 100 kHz (-8.0 kOe/K) (Ref. 9) in the same orientation. We also observe that T_Q is independent of resonant mode (500 Hz–50 kHz) within an uncertainty of 0.1 K. This implies that the pinning time scale (see below) changes from $\tau > 1$ ms to $\tau < 3$ μs over a temperature interval of less than 0.1 K.

We have observed mode-dependent structure in $\Delta f/f$ and $1/Q$, as well as complex field and temperature hysteresis effects in $NbSe_2$; a more complete discussion of these data will be given in a future publication.

Data for the temperature dependences of R, $\Delta f/f$, and $1/Q$ for the first overtone (f = 3.1 kHz) of a Nb reed measured in the two sample and field orientations are shown in Figs. 2 and 3. T_Q lies *within* the resistive transition region (below a high-temperature tail) for the parallel orientation shown in Fig. 2. It is noteworthy that the onsets of the $1/Q$ and $\Delta f/f$ anomalies lie below a kink (labeled by T_k) in the temperature dependence of the resistance that we associate with a transition to surface superconductivity (H_{c3}),[19] as discussed below. This is consistent with the unusual field broadening of the transition *above* the resistive midpoint, and with elastic properties such as $1/Q$ and $\Delta f/f$ only being sensitive to bulk changes in the sample that occur below T_k. These results are confirmed for all fields and modes measured in the parallel orientation.

On the other hand, we find that T_Q lies considerably below T_R in the perpendicular orientation, where T_R is the temperature at which the resistance approximately reaches zero, as shown in Fig. 3. We note that this result

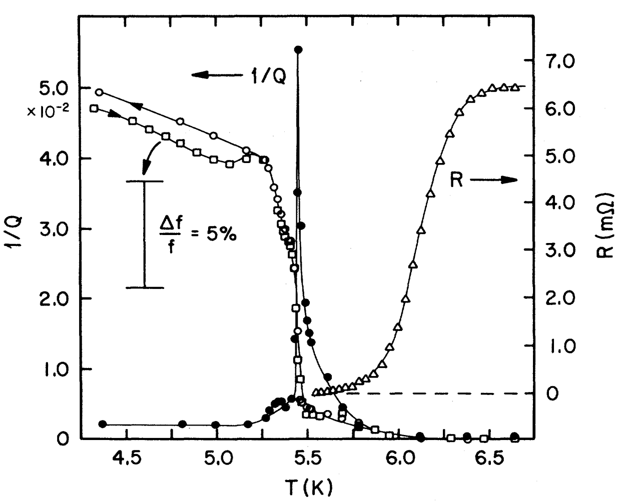

FIG. 1. Resistance R (triangles), reciprocal quality factor $1/Q$ (solid circles), and frequency shift $\Delta f/f$ (open circles: cooling; squares: warming) vs temperature T for a $NbSe_2$ single-crystal reed with the *ab* planes and long dimension perpendicular to an applied field H = 11 kOe. Elastic measurements were made on the fundamental resonance at 680 Hz, and R was determined with a 100 μA current. The lines are a guide to the eye.

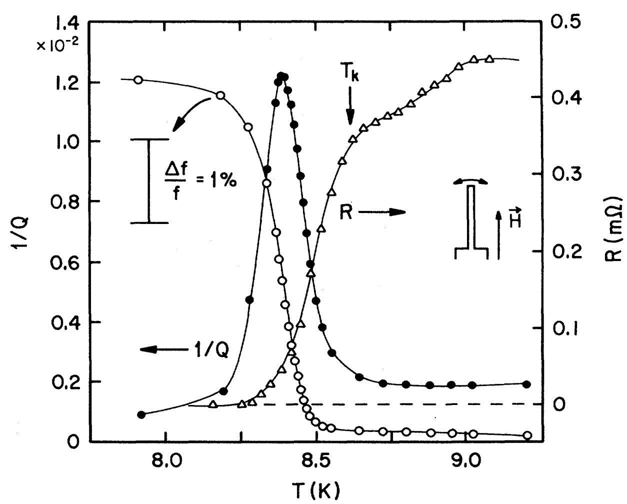

FIG. 2. Resistance R (triangles), reciprocal quality factor $1/Q$ (solid circles), and frequency shift $\Delta f/f$ (open circles: cooling) vs temperature T for a Nb foil reed mounted with the long dimension parallel (see inset) to an applied field H = 0.92 kOe. Elastic measurements were made on the first overtone at 3.1 kHz, and R was determined with a 1 mA current. The lines are a guide to the eye.

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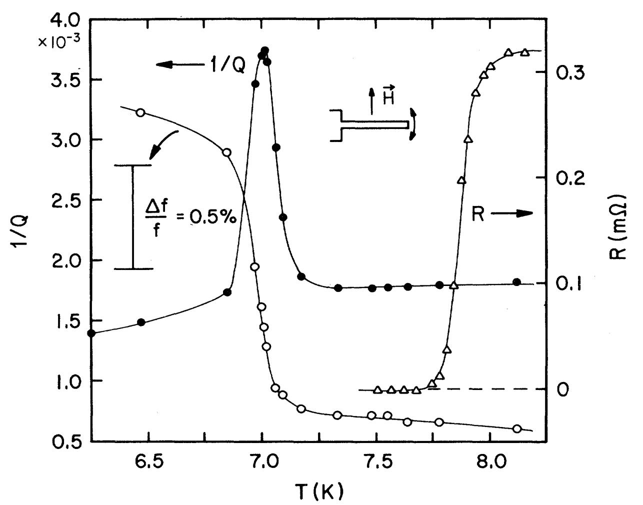

FIG. 3. Resistance R (triangles), reciprocal quality factor $1/Q$ (solid circles), and frequency shift $\Delta f/f$ (open circles: cooling) vs temperature T for a Nb foil reed mounted with the long dimension perpendicular (see inset) to an applied field $H = 1.83$ kOe. Elastic measurements were made on the first overtone at 3.1 kHz, and R was determined with a 1 mA current. The lines are a guide to the eye.

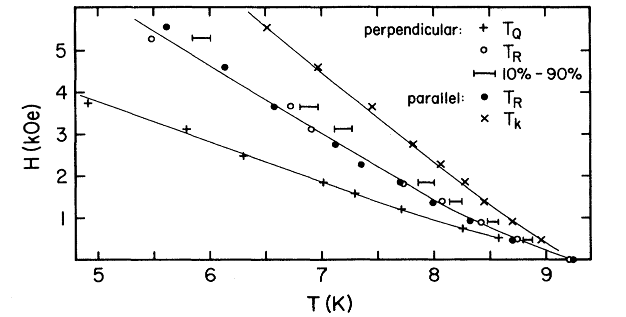

FIG. 4. Applied magnetic field H vs temperature T, showing the field dependences of the experimental temperatures defined by a peak in the reciprocal quality factor (T_Q) and the attainment of zero resistance (T_R) for the first overtone (3.1 kHz) of a Nb reed in the perpendicular orientation. Data for the parallel orientation include T_R and the temperature of the kink in the electrical resistance (T_k). Resistances were measured with a 100 μA current, horizontal bars represent the 10%–90% resistive transition widths, and lines are guides to the eye. T_Q data for the parallel orientation are not shown, but are very close to T_R ($T_R - T_Q < 0.1$ K). The resistance kink is not observed in the perpendicular orientation.

does not appear to be limited by our resistance resolution, as a factor of 10 change in measuring current only shifts T_R by <0.1 K. For example, at $H = 1.83$ kOe, $T_R - T_Q \approx 0.7$ K, independent of mode frequency (0.7–11 kHz).

In Fig. 4, we plot the field dependence of T_Q, T_R, and the resistive transition width (10% to 90% values) for the perpendicular orientation; and T_R and T_k are plotted for the parallel orientation. Note that $H(T_k) \approx (1.7) \times H(T_R)$, and that there is no field broadening *above* the midpoint of the resistive transition in the perpendicular geometry, corroborating the identification of $T_k(H)$ with the boundary between the bulk vortex state and surface superconductivity.

Brandt[11–13] has analyzed the behavior of superconducting reeds vibrating in an applied magnetic field; in particular, he has calculated changes in resonant frequency and damping for extreme type-II materials in external fields $H > 2H_{c1}$, the lower critical field, so that Meissner currents (demagnetization effects) can be neglected.

When the FL are rigidly pinned, the frequency should increase rapidly at T_c, primarily due to the energy needed to bend field lines through (around) the reed oriented perpendicular[13] (parallel[11]) to the applied field. Because there are larger surface screening currents that interact with the applied field in the parallel orientation, $\Delta f/f$ is a factor of w/d larger than for the perpendicular orientation,[13,14] where w and d are the width and thickness of the reed (i.e., an order of magnitude increase for our samples). Brandt predicts $\Delta f \propto H^2$ (for $\Delta f/f \ll 1$), and that Δf is approximately independent of frequency (mode) for both orientations.[11] An amplitude-dependent damping peak is predicted at T_c (i.e., the equilibrium transition temperature) as the pinning vanishes upon entry into the normal state.

On the other hand, the magnitude of $\Delta f/f$ will decrease in the case of nonrigid pinning (finite Labusch parameter[20]).[12] If the FL depinning is thermally activated, the frequency shift and internal friction peak will shift to a lower temperature where the thermally activated diffusion constant $D(T) \approx fl^2$, where l is an effective FL diffusion length. For the fundamental resonance in the parallel orientation, $l \approx L$, the length of the reed, whereas for the perpendicular, $l \approx d \ll L$ (Ref. 13), possibly explaining the experimental depression of T_Q below T_R.

The Ginzburg-Landau $\kappa \approx 9$ and $H_{c1}(0) < 400$ Oe for $NbSe_2$ in the perpendicular orientation (Ref. 16), so we should be far enough into the vortex state for $H > 1$ kOe for a comparison to Brandt's theory to be feasible. However, in the case of Nb it is not clear if we are in the low magnetization limit since κ is not expected to be large. Nevertheless, the same value of T_R is observed for both orientations of Nb reeds (see Fig. 4), indicating that demagnetization effects are not large for this particular sample.

We find that, for all of our samples, $\Delta f/f$ for the parallel orientation in a given field is an order of magnitude larger than for the perpendicular orientation, as expected for nearly rigid pinning. For the $NbSe_2$ samples, Δf is approximately independent of frequency, but the field dependence is typically weaker than quadratic: e.g., $\Delta f \propto H^m$, with $1 < m < 1.8$, for both orientations, in conflict with Brandt's predictions. For the Nb sample, Δf increases slowly with f; an H^2 dependence is observed for the parallel orientation, and an approximate $H^{3/2}$ dependence for the perpendicular.

Such an $H^{3/2}$ power law has also been observed[14] for suspensions of small superconducting particles, for which no substantial bending of field lines around the particles is expected. In this case, Δf can be shown to reflect only the elastic forces on the FL and is a direct measure of the Labusch parameter $\alpha \propto H^{3/2}$, rather than the magnetic-field energy (FL tilt modulus $\propto H^2$), as in the strong-pinning, zero-magnetization case considered in Ref. 1. For this explanation to apply to our own data for perpendicular reed orientations, the Labusch parameter must satisfy $\alpha < \mu_0(H/d)^2 \approx 3.2\times10^{12} N/m^4$ at $H=1$ kOe (Ref. 14). α cannot be much less than this in order to account for the 1 order-of-magnitude difference in $\Delta f/f$ between the two orientations in Nb. Further experimentation will investigate this possibility.

Finally, we consider the observed lack of frequency dependence in T_Q. The effective diffusion length is the distance between nodes and antinodes of the flexural resonance for the parallel orientation; i.e., $l \approx L/n$, where n is the mode number.[13,21] However, the resonant frequency $f(H=0) \propto n^2$ (Ref. 22), so that $D(T)$ and T_Q should be independent of mode. In contrast, for the perpendicular orientation, $l=d$ for all modes,[13] and we expect T_Q to increase with frequency, in conflict with our present data.

In conclusion, our results are consistent with those of SGXW,[10] and show that an extended reversible region may be a general feature of thin type-II superconductors aligned with long dimension perpendicular to an applied magnetic field. Surprisingly, the reversible region is wider in the Nb sample than for the $NbSe_2$, which is both more anisotropic and has a much smaller coherence length ($\xi_{0c} \approx 26$ Å) and larger κ than Nb.[8,9,16] Present interpretations of data that treat extended regions of reversibility exclusively in terms of small coherence length or high-T_c mechanisms should be treated with caution.

We wish to thank Dr. E. H. Brandt for very informative and stimulating discussions, and Dr. X.-D. Xiang for help in the design of the vibrating reed apparatus. We have also benefited from discussions with Professor J. Clem, Dr. L. Campbell, and Dr. L. Daemon. This work was funded in part by National Science Foundation Grant No. DMR-89-15440, and by the Research Corporation.

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